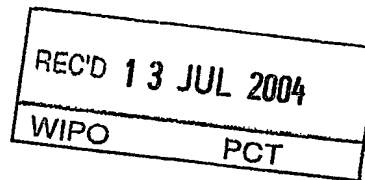




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Dated

1 July 2004

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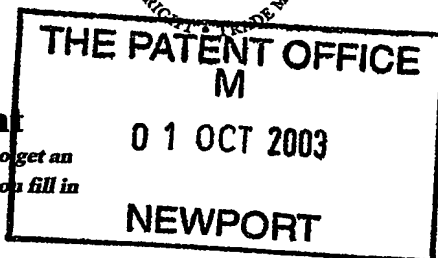


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P01/7700 000-0322963.0

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Request for grant of a patent

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The Patent Office

Cardiff Road
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South Wales
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1. Your reference

AA 1676 GB

2. Patent application number

(The Patent Office will fill in this part)

0322963.0

23 JUN 2003

3. Full name, address and postcode of the or of each applicant (underline all surnames)

JOHNSON MATTHEY PUBLIC LIMITED COMPANY
2-4 COCKSPUR STREET
TRAFALGAR SQUARE
LONDON SW1 5BQ

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

GB 536 268 007

4. Title of the invention

METHODS OF CONTROLLING REDUCANT
ADDITION

5. Name of your agent (if you have one)

ANDREW DOMINIC NUNN

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

JOHNSON MATTHEY TECHNOLOGY CENTRE
BLOUNTS COURT
SONNING COMMON
READING RG4 9NH

Patents ADP number (if you know it)

3991411501

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number
(if you know it)

Date of filing
(day / month / year)

GB

0314463.1

23 JUNE 2003

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

YES

- a) any applicant named in part 3 is not an inventor, or
 - b) there is an inventor who is not named as an applicant, or
 - c) any named applicant is a corporate body.
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Patents Form 1/77

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Continuation sheets of this form

Description

Claim(s)

Abstract

Drawing(s)

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3

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Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (*Patents Form 7/77*)

Request for preliminary examination and search (*Patents Form 9/77*)

Request for substantive examination (*Patents Form 10/77*)

Any other documents
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11.

I/We request the grant of a patent on the basis of this application.

Signature

F E Wishart
I C WISHART

Date

30 Sep 03

12. Name and daytime telephone number of person to contact in the United Kingdom

MRS F E STRANGE 0118 924 2125

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METHODS OF CONTROLLING REDUCANT ADDITION

The present invention relates to methods of controlling reductant addition to catalytically convert NO_x in exhaust gas of an internal combustion engine to N_2 .

5

It is known to catalytically reduce NO_x in exhaust gas of an internal combustion engine to N_2 with a suitable reductant. Three examples are selective catalytic reduction (SCR), lean- NO_x catalysis and NO_x -trap regeneration.

10

In SCR, the reductant is typically a NO_x -specific reactant. By " NO_x specific reactant" herein, we mean a reducing agent that, in most conditions, preferentially reduces NO_x over other components of a gaseous mixture. Examples of NO_x -specific reactants include nitrogenous compounds such as nitrogen hydrides, e.g. ammonia (NH_3) or hydrazine, or an NH_3 precursor.

15

By " NH_3 precursor" we mean one or more compounds from which NH_3 can be derived, e.g. by hydrolysis. These include urea ($\text{CO}(\text{NH}_2)_2$) as an aqueous solution or as a solid or ammonium carbamate ($\text{NH}_2\text{COONH}_4$). If the urea is used as an aqueous solution, a eutectic mixture, e.g. 32.5% urea (aq), is preferred. Additives can be included

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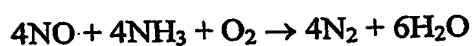
in the aqueous solutions to reduce the crystallisation temperature.

25

Known SCR catalysts include Pt-based catalysts, which can catalyse the reduction of NO_x with NH_3 at between about 175°C and about 250°C , medium temperature vanadium-based catalysts e.g. $\text{V}_2\text{O}_5/\text{TiO}_2$, which operate in the temperature range between about 260°C and about 450°C and zeolite-based catalysts which function with increasing activity at increasing temperature.

30

Several chemical reactions occur in the NH_3 SCR system, all of which represent desirable reactions which reduce NO_x to elemental nitrogen. The dominant reaction mechanism is represented in equation (1).



(1)

Competing, non-selective reactions with oxygen can produce secondary emissions or may unproductively consume NH_3 . One such non-selective reaction is the complete oxidation of NH_3 , represented in equation (2).



Urea hydrolyses at temperatures above 160°C according to equation (3) to liberate NH_3 itself. It also thermally decomposes at this temperature and above according to equations (4) and (5) resulting in reduction of NO_x .

10



15 It will be appreciated that at lower temperatures, below about $100\text{-}200^\circ\text{C}$, NH_3 can also react with NO_2 to produce explosive mixture of ammonium nitrate (NH_4NO_3) and ammonium nitrite (NH_4NO_2) according to equation (6).



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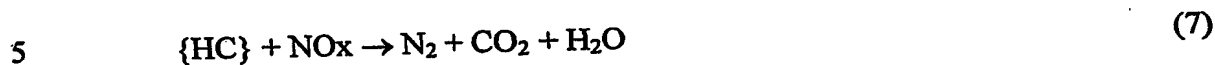
For the avoidance of doubt, the present invention does not embrace such reactions or the promotion of conditions which bring them about. For example, the reaction can be avoided by ensuring that the temperature does not fall below about 200°C or by supplying into a gas stream less than the precise amount of NH_3 necessary for the stoichiometric reaction with NO_x (1 to 1 mole ratio).

25

Lean- NO_x catalysts (LNCs) are sometimes also referred to in the literature as non-selective catalytic reduction (NSCR) catalysts, hydrocarbon selective catalytic reduction (HC-SCR) catalysts, lean- NO_x reduction catalysts, "DeNO $_x$ catalysts" and

30 NO_x occluding catalysts.

In lean- NO_x catalysis, hydrocarbons (HC) react with nitrogen oxides (NO_x), rather than oxygen (O_2), to form nitrogen (N_2), carbon dioxide (CO_2) and water (H_2O) according to equation (7).

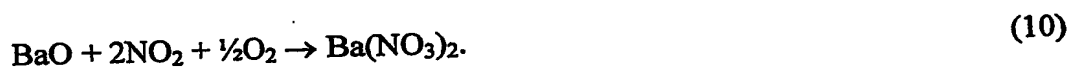


The competitive, non-selective reaction with oxygen is given by Equation (8).



There are two preferred groups of LNC to selectively promote the desired reaction (7) described in the literature: platinum (Pt) on alumina (Al_2O_3) and copper (Cu)-substituted zeolite such as Cu/ZSM-5.

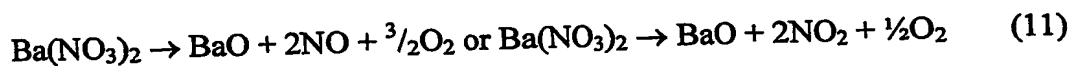
A typical NO_x -trap formulation includes a catalytic oxidation component, such as platinum, a NO_x -storage component, such as compounds of alkali metals e.g. potassium and/or caesium; compounds of alkaline earth metals, such as barium or strontium; and compounds of rare-earth metals, typically lanthanum and/or yttrium; and a reduction catalyst, e.g. rhodium. One mechanism commonly given for NO_x -storage during lean engine operation for this formulation is set out in equation (9) and (10) using barium as an illustrative Example.



In step (9), the nitric oxide reacts with oxygen on active oxidation sites on the platinum to form NO_2 . The second step involves adsorption of the NO_2 by the storage material in the form of an inorganic nitrate.

When the engine runs intermittently under rich conditions or at elevated temperatures, the nitrate species become thermodynamically unstable and decompose, producing NO or NO_2 according to equation (11) below. Under rich conditions, these

nitrogen oxides are subsequently reduced by carbon monoxide, hydrogen and hydrocarbons to N_2 , which can take place over the reduction catalyst.



In the reactions of (9)-(12) above, the reactive barium species is given as the oxide. However, it is understood that in the presence of air or exhaust gas containing CO_2 and H_2O the barium may also be in the form of the carbonate or possibly the hydroxide. The above reaction schemes can be adapted accordingly for species of barium other than the oxide. We also explain in our WO 00/21647 that NO_x -specific reactants can be used to regenerate a NO_x -trap.

A problem with the above NO_x reduction methods is to control the addition of the reductant. If too little reductant is added, NO_x reduction may be inadequate to meet an emission standard. If too much reductant is added this can cause a number of problems. For example, if the reductant is ammonia, its release into the atmosphere is undesirable because it is a biological poison and it has an unpleasant odour. Whilst excess ammonia can be oxidised using a suitable catalyst downstream of the NO_x -reduction catalyst, this produces NO_x , thus defeating the very purpose of the NO_x reduction method. Hydrocarbon fuels, e.g. diesel or gasoline, are also legislated components of exhaust gas and so emission of excess hydrocarbon reductant can cause the system to fail a relevant emission standard.

Systems to control reductant addition are known, but tend to require very complicated control regimes involving multiple sensor inputs and processors to run complex algorithms. As a result, such systems are very expensive.

We have investigated methods of calibrating reductant addition and of controlling reductant addition by feedback. We have now devised a number of simple methods and systems that are cheap and effective at reducing reductant-based emissions. Systems embodying these methods are particularly relevant to the retrofit market.

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A first aspect of the invention makes use of only a single temperature measurement, e.g. using a thermocouple, as the control input. Preferably, no reductant is added below a pre-determined temperature because the temperature is too low for the complete NO_x removal reaction to take place. As the load on an engine is increased, the NO_x level in the exhaust gas increases in a roughly linear fashion. Similarly, the exhaust gas temperature increases with load. We expect, therefore, that for a given engine in a particular application there is a relationship between NO_x content in the exhaust gas and its temperature. And, that this may approximate to a linear relationship. So, in its simplest form, in this aspect of the invention, the rate of reductant introduced is proportional to the temperature of the exhaust gas. Addition of a reductant in this way provides a very efficient and simple way of maintaining NO_x reduction on a vehicle. The actual amount of reductant added at different exhaust gas temperatures can depend on the nature of the specific application. But, there may be a trend for more reductant to be needed to remove larger amounts of NO_x at higher temperatures. In practice a calibration process would determine the exact amounts.

In a system embodiment of the first aspect, the invention provides an exhaust system for an internal combustion engine, which system comprising a catalyst for reducing NO_x in exhaust gas to N_2 with a suitable reductant, a source of reductant, means for contacting the catalyst with the reductant, means for sensing the temperature of the exhaust gas and/or the catalyst bed and means, when in use, for controlling reductant addition, wherein the reductant addition control means supplies an amount of reductant to the catalyst corresponding to a measured temperature value of the exhaust gas and/or catalyst bed, which temperature value has been pre-determined to correlate, in use, with an amount of NO_x in the exhaust gas.

In a method embodiment of the first aspect, the invention provides a method of calibrating an amount of reductant addition required to reduce NO_x in an exhaust gas of an internal combustion engine to N_2 over a suitable catalyst, which method comprising measuring NO_x in the exhaust gas at a plurality of exhaust gas and/or catalyst bed temperatures and correlating each exhaust gas and/or catalyst bed temperature value with an amount of reductant addition required to reduce the NO_x over the catalyst.

A second aspect of the invention provides a simple means of controlling reductant addition by feedback in real-time. In the second aspect, an oxidation catalyst is disposed downstream of the NO_x removal catalyst which is contacted with a reductant. The system is arranged so that the gas is always lean over the oxidation catalyst, e.g. by using secondary addition of air if required. As in the first aspect described above, desirably no reductant is added below a certain critical exhaust gas temperature, at which NO_x reduction is ineffective. Above this temperature, increasing the amount of reductant causes increasing amounts of NO_x in the exhaust gas to be reduced. In practice there is a limit to this effect, and then introducing more reductant fails to enhance NO_x reduction. So, there is a region of small excess reductant slip from the NO_x reduction catalyst that corresponds to a desirable addition rate above which any more reductant is wasted.

Because the overall gas composition is arranged to be lean, any excess reductant can be oxidised over the downstream oxidation catalyst, and the resulting exotherm may result in a temperature increase across the oxidation catalyst. The inlet temperature to the oxidation catalyst may vary considerably during use, but in this method we are concerned only with the ΔT that is a measurement of the excess reductant present. The control strategy is based in adjusting the rate of reductant addition to keep the measured ΔT within a pre-determined range corresponding to an optimum NO_x removal: reductant addition is increased if ΔT is too small, or decreased if ΔT is larger than desired for optimum efficient NO_x conversion.

According to a system embodiment of the second aspect of the invention, there is provided an exhaust system for an internal combustion engine, which system comprising a catalyst for reducing NO_x in an exhaust gas to N_2 with a suitable reductant, a source of reductant, means for contacting the catalyst with the reductant, an oxidation catalyst disposed downstream of the NO_x reduction catalyst, means for determining a temperature difference (ΔT) across the oxidation catalyst and means, when in use, for controlling reductant addition, wherein the reductant addition control means controls a rate of reductant addition to maintain ΔT within a pre-determined range, wherein the system is configured so that the exhaust gas composition over the oxidation catalyst is lean.

In a method embodiment of the second aspect of the invention, a method of controlling, by feedback, addition of reductant to a catalyst suitable for reducing NO_x in an exhaust gas of an internal combustion engine to N_2 with the reductant comprises providing an oxidation catalyst for oxidising the reductant downstream of the NO_x reduction catalyst, measuring the exhaust gas temperature upstream of the oxidation catalyst, measuring the exhaust gas temperature downstream of the oxidation catalyst, determining the difference between the inlet and the outlet temperatures (ΔT) and adjusting reductant addition so that ΔT is within a pre-determined range.

10 The internal combustion engine can be a diesel or lean-burn gasoline engine, such as a gasoline direct injection engine. The diesel engine can be a light duty engine or a heavy-duty engine, as defined by the relevant legislation.

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CLAIMS:

1. An exhaust system for an internal combustion engine, which system comprising a catalyst for reducing NO_x in exhaust gas to N_2 with a suitable reductant, a source of
5 reductant, means for contacting the catalyst with the reductant, means for sensing the temperature of the exhaust gas and/or the catalyst bed and means, when in use, for controlling reductant addition, wherein the reductant addition control means supplies an amount of reductant to the catalyst corresponding to a measured temperature value of the exhaust gas and/or catalyst bed, which temperature value has been pre-determined to
10 correlate, in use, with an amount of NO_x in the exhaust gas.
2. A method of calibrating an amount of reductant addition required to reduce NO_x in an exhaust gas of an internal combustion engine to N_2 over a suitable catalyst, which method comprising measuring NO_x in the exhaust gas at a plurality of exhaust gas and/or
15 catalyst bed temperatures and correlating each exhaust gas and/or catalyst bed temperature point with an amount of reductant addition required to reduce the NO_x over the catalyst.
3. An exhaust system for an internal combustion engine, which system comprising a
20 catalyst for reducing NO_x in exhaust gas to N_2 with a suitable reductant, a source of reductant, means for contacting the catalyst with the reductant, an oxidation catalyst disposed downstream of the NO_x reduction catalyst, means for determining a temperature difference (ΔT) across the oxidation catalyst and means, when in use, for
controlling reductant addition, wherein the reductant addition control means controls a
25 rate of reductant addition to maintain ΔT within a pre-determined range, wherein the system is configured so that the exhaust gas composition over the oxidation catalyst is lean.
4. A method of controlling, by feedback, addition of reductant to a catalyst suitable
30 for reducing NO_x in an exhaust gas of an internal combustion engine to N_2 with the reductant, which method comprising providing an oxidation catalyst for oxidising the reductant downstream of the NO_x reduction catalyst, measuring the exhaust gas temperature upstream of the oxidation catalyst, measuring the exhaust gas temperature

downstream of the oxidation catalyst, determining the difference between the inlet and the outlet temperatures (ΔT) and adjusting reductant addition so that ΔT is within a pre-determined range.

- 5 5. An exhaust system according to claim 1 or 3, wherein reductant addition is suspended when the exhaust gas and/or the catalyst bed temperature is below a pre-determined temperature.
6. An exhaust system according to claim 1, 3 or 5, wherein reductant addition is
10 suspended when the exhaust gas and/or the catalyst bed temperature is above a pre-determined temperature.
7. A method according to claim 2 or 4, wherein reductant addition is suspended
15 when the exhaust gas and/or the catalyst bed temperature is below a pre-determined temperature.
8. A method according to claim 2, 4 or 7, wherein reductant addition is suspended
when the exhaust gas and/or the catalyst bed temperature is above a pre-determined
20 temperature.
9. An exhaust system according to claim 3, wherein the rate of reductant addition is
decreased if ΔT is too large.
10. A method according to claim 4, wherein the rate of reductant addition is
25 decreased if ΔT is too large.
11. An exhaust system according to claim 1, 3, 5, 6 or 9, wherein the reductant is a hydrocarbon and the catalyst is a lean- NO_x catalyst.
- 30 12. An exhaust system according to claim 1, 3, 5, 6 or 9, wherein the reductant is a NO_x -specific reactant and the catalyst is a selective catalytic reduction (SCR) catalyst.

13. An exhaust system according to claim 11 or 12, wherein the catalyst comprises a NO_x-absorbent.
14. A method according to claim 2, 4, 7, 8 or 10, wherein the reductant is a hydrocarbon and the catalyst is a lean-NO_x catalyst.
15. A method according to claim 2, 4, 7, 8 or 10, wherein the reductant is a NO_x-specific reactant and the catalyst is a selective catalytic reduction (SCR) catalyst.
16. A method according to claim 14 or 15, wherein the catalyst comprises a NO_x-absorbent.

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